

Quantum-State Purity of Heralded Single Photons Produced from Frequency-Anti-Correlated Biphotons

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We analyze the quantum-state purity of heralded single photons produced from frequency-anti-correlated biphotons. We find that the quantum-state purity in time-frequency domain depends strongly on the response time uncertainty of the trigger-photon detector that heralds the generation of its paired photon. If the trigger response time is much shorter than the two-photon coherence time, the time-frequency quantum-state purity of heralded single photons approaches unity and the heralded single photon is in a nearly pure state. If the trigger response time is much longer than the two-photon coherence time, the heralded photon is then projected onto a mixed state. Making use of the time-frequency entanglement, heralded single photons with a well-defined temporal wave function or a frequency superposition state can be produced and engineered. This time-frequency entanglement allows for shaping heralded single photons through nonlocal spectral modulation.

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Correlated photon pairs can be used to generate heralded single photons: the detection of one photon heralds the presence of the remaining one and projects it onto a single-photon Fock state. Spontaneous parametric down conversion (SPDC, $\chi^{(2)}$ nonlinear process) [1, 2] and spontaneous four-wave mixing (SFWM, $\chi^{(3)}$ nonlinear process) [3, 4] have been two standard methods for producing paired photons. As driven by continuous-wave (CW) pump laser fields, the photon pairs generated from these parametric processes are time-frequency entangled because of the energy conservation raised from the time-translation symmetry, *i.e.*, the sum of the frequencies of paired two photons is fixed while individual photons have their bandwidths. It has been commonly believed that, this frequency anti-correlation would project the heralded single photon into a mixture of frequency modes in its bandwidth [5, 7], and spectral filtering to obtain an approximate pure single-photon state results in a dramatically reduced photon rate by discarding photons outside of the filter frequency mode [8, 9]. A complete different approach to obtain a heralded pure single-photon state is thus eliminating the entanglement and generating photon pairs with factorable (frequency-uncorrelated) joint spectrum driven by pulsed lasers [5–7, 10–15].

For frequency-anti-correlated biphotons, the heralding detection is commonly modeled as tracing over the trigger photon and thus reduces the heralded photon into a mixed state [6]. In this Letter, we point out that an ideal heralding process with an instantaneous trigger-photon detection projects the remaining frequency-anti-correlated photon onto a pure quantum state, which is a superposition state of its frequency components. When the trigger photon detection has a finite response time, it degrades the purity of the single photons. We analyze the quantum-state purity of heralded single photons produced from frequency-anti-correlated biphotons. If the trigger response time is much longer than the two-photon

coherence time, the heralded photon is projected onto a mixed state, as well known for decades. If the trigger response time is much shorter than the two-photon coherence time, we find that, the time-frequency quantum-state purity of heralded single photons approaches nearly unity without any need of spectral filtering.

Let's start with the biphoton state (in time-frequency space) of a frequency anti-correlated photon pair

$$|\Psi_{12}\rangle = \int d\Omega \Phi(\Omega) \hat{a}_2^\dagger(\omega_{20} - \Omega) \hat{a}_1^\dagger(\omega_{10} + \Omega) |0\rangle, \quad (1)$$

where $|0\rangle$ is the vacuum state, ω_{10} and ω_{20} are the central angular frequencies of photons 1 and 2. \hat{a}_1^\dagger and \hat{a}_2^\dagger are the field creation operators. $\Phi(\Omega)$ is the two-photon joined spectrum function. The frequency entanglement of the biphoton state is a result of the energy conservation $\omega_1 + \omega_2 = \omega_{10} + \omega_{20}$. The field operators in time domain can be expressed as:

$$\hat{a}_i(t) = \frac{1}{\sqrt{2\pi}} \int d\omega \hat{a}_i(\omega) e^{-i\omega t}. \quad (2)$$

The field operators satisfy the commutation relations $[\hat{a}_i(\omega), \hat{a}_j^\dagger(\omega')] = \delta_{ij} \delta(\omega - \omega')$ and $[\hat{a}_i(t), \hat{a}_j^\dagger(t')] = \delta_{ij} \delta(t - t')$. In the state described in Eq. (1), the two photons are perfectly paired. It can be shown that the photon pair generation rate R and the individual single-photon rates R_i are equal and time invariant:

$$R = R_i = \langle \Psi_{12} | \hat{a}_i^\dagger(t) \hat{a}_i(t) | \Psi_{12} \rangle = \frac{1}{2\pi} \int d\Omega |\Phi(\Omega)|^2, \quad (3)$$

because the system is driven by CW pump fields and has the time-translation symmetry.

Detection of the photon 2 at time t_2 reduces the two-photon state in Eq. (1) to the following state:

$$|\Psi_1\rangle_2 = \frac{1}{\sqrt{R}} \hat{a}_2(t_2) |\Psi_{12}\rangle, \quad (4)$$

which is the heralded single-photon state with the normalization factor $1/\sqrt{R}$. To prove this is a pure quantum state, we look at its density operator

$$\hat{\rho}_{1|2} = |\Psi_1\rangle_{22}\langle\Psi_1|. \quad (5)$$

It is obvious that

$$\hat{\rho}_{1|2}^2 = \hat{\rho}_{1|2}, \quad (6)$$

which shows the heralded single-photon state in Eq. (4) is indeed a pure quantum state. Therefore, we have proved that in the ideal heralding process described above, where the detection of the trigger photon 2 has an infinitely short response time and there is no time uncertainty in the heralding process, the projected single photon is in a pure state.

Now we turn to the real situation where the trigger-photon detector has a finite response time Δt : one does not know the exact time origin of the heralded single photon during this response time window. In this case, the density operator can be expressed as

$$\begin{aligned} \hat{\rho}_{1|2} &= \frac{1}{\Delta t} \int_{-\Delta t/2}^{\Delta t/2} dt_2 |\Psi_1\rangle_{22}\langle\Psi_1| \\ &= \frac{1}{R\Delta t} \int_{-\Delta t/2}^{\Delta t/2} dt_2 \hat{a}_2(t_2) |\Psi_{12}\rangle \langle\Psi_{12}| \hat{a}_2^\dagger(t_2). \end{aligned} \quad (7)$$

Making use of Eq. (2), the density operator can be rewritten as

$$\begin{aligned} \hat{\rho}_{1|2} &= \frac{1}{2\pi R} \int d\omega_2 d\omega'_2 \text{sinc}\left[\frac{(\omega_2 - \omega'_2)\Delta t}{2}\right] \\ &\quad \times \hat{a}_2(\omega_2) |\Psi_{12}\rangle \langle\Psi_{12}| \hat{a}_2^\dagger(\omega'_2). \end{aligned} \quad (8)$$

We then get the density matrix element in the frequency domain

$$\begin{aligned} \bar{\rho}_{1|2}(\omega_1, \omega'_1) &= \langle 0 | \hat{a}_1(\omega_1) \hat{\rho}_{1|2} \hat{a}_1^\dagger(\omega'_1) | 0 \rangle \\ &= \frac{1}{2\pi R} \text{sinc}\left[\frac{(\omega_1 - \omega'_1)\Delta t}{2}\right] \Phi(\omega_1 - \omega_{10}) \Phi^*(\omega'_1 - \omega_{10}) \end{aligned} \quad (9)$$

The quantum-state purity of the heralded single photons can be computed from $\gamma = \text{Tr}(\hat{\rho}_{1|2}^2)$ [6, 16]. With the frequency-entangled biphoton state in Eq. (1), we obtain the purity of its heralded single-photon state

$$\begin{aligned} \gamma &= \frac{1}{(2\pi R)^2} \int d\Omega d\Omega' \text{sinc}^2\left[\frac{(\Omega - \Omega')\Delta t}{2}\right] \\ &\quad \times |\Phi(\Omega)\Phi(\Omega')|^2 \end{aligned} \quad (10)$$

Obviously, as the trigger-photon detection is instantaneously fast ($\Delta t \rightarrow 0$), the density operator is reduced to that of a pure state $\hat{\rho}_{1|2} \rightarrow \hat{\rho}_{1|2}(t_2 = 0)$ and we have an unity purity ($\gamma = 1$). This is consistent with our previously described ideal heralding process that projects the remaining photon onto a pure quantum state. As the trigger-photon detector is ultraly slow ($\Delta t \rightarrow \infty$), $\text{sinc}\left[\frac{(\omega_1 - \omega'_1)\Delta t}{2}\right] \rightarrow 2\pi\delta(\omega_1 - \omega'_1)/\Delta t$, all non-diagonal

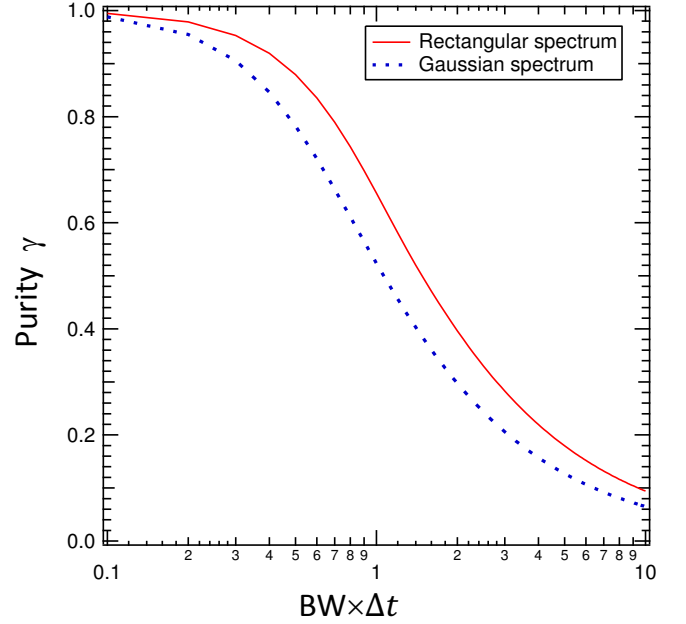


FIG. 1. (color online). Quantum-state purity γ as a function of $BW \times \Delta t$, the product of biphoton bandwidth (BW) and the trigger-photon detection response time (Δt). The solid (red) curve is calculated from the rectangular-shape spectrum, and the dotted (blue) curve from the Gaussian-shape spectrum with BW as the full width at half maximum.

density matrix elements in Eq. (9) vanishes and the heralded single photon is in a completely mixed state.

For the case with a finite Δt , the quantum-state purity is characterized by Eq. (10). As the trigger-photon detection response time is much shorter than the two-photon coherence time (inverse of the bandwidth of the joint spectrum) such that $(\Omega - \Omega')\Delta t \ll 1$ holds within the bandwidth, we have $\gamma \sim 1$ and the heralded photon is in a nearly pure state. To show how the purity of the heralded single photon state is affected by the finite Δt , we consider two examples in the following.

Case 1 with a rectangular-shape spectrum. In the first, we consider transform-limited biphotons with maximum frequency entanglement within their angular-frequency bandwidth of $2\pi BW$. In this case, the joint spectrum function $\Phi(\Omega) = \sqrt{R/BW}$ is nonzero only within the bandwidth $\Omega \in [-\pi BW, \pi BW]$. The solid curve in Fig. 1 shows the numerical result of the purity as a function of the trigger response time. At $BW\Delta t \leq 0.1$, the purity $\gamma \geq 0.99$. The purity decreases as we increase the response time, but not very badly. As the response time approaches the two-photon coherence time ($BW\Delta t = 1$), the purity is still as high as 0.66. As we further increase the response time to make $BW\Delta t = 10$, the purity drops down to 0.09.

Case 2 with a Gaussian-shape spectrum. In this case, the joint spectrum function can be expressed as $\Phi(\Omega) =$

$\sqrt{\frac{4R\sqrt{\pi\ln 2}}{2\pi BW}}e^{-2\ln 2(\Omega/2\pi BW)^2}$, where BW is the full width at half maximum of $|\Phi(\Omega)|^2$. The numerical result of the purity as a function of the trigger response time is plotted as the dotted curve in Fig. 1, which is comparable to that of Case 1 with a rectangular-shape spectrum. At $BW\Delta t = 0.1$, the purity $\gamma = 0.99$. At $BW\Delta t = 1$, the purity becomes 0.52. The purity drops down to 0.07 as we further increase the response time to make $BW\Delta t = 10$.

In both cases, the quantum-state purity $\gamma > 0.98$ at $BW\Delta t = 0.1$, and holds well above 0.90 for $BW\Delta t < 0.3$. We can treat such heralded photon as a pure single-photon state. For biphotons generated from SPDC or SFWM in solid-state materials without spectral filtering or cavity enhancement, their bandwidths are normally much wider than THz. As a result, it is impossible to heralding pure single photons from such frequency-entangled photon pair source using a commercially available single-photon detector with a typical time resolution of about 1 ns. Even the state-of-art single-photon detector with an ultra-high time resolution of about 20 ps is not fast enough. Therefore, disentangling the photon pairs into a factorable joint spectrum by shaping the pump field temporal modes has been considered as the only achievable method to produce heralded pure single-photon states from these wide-band sources without spectral filtering. This situation has been changed recently with the development of narrowband biphotons generation from SFWM in cold atoms [4, 17–20] and cavity-enhanced SPDC [21, 22]. These biphotons, having bandwidths ranging from below 100 MHz down to sub-MHz and coherence time from 10 ns up to more than 1 μ s, are ideal for generating heralded pure single-photon states using commercial detectors.

As the time origin is settled by the detection of the trigger photon ($t_2 = 0$), the heralded photon has a well-defined temporal wave function. To illustrate this, we work at $BW\Delta t \leq 1$ and thus it can be treated as an ideal heralding process. From Eq. (4), we obtain the temporal wave function of the heralded single photon:

$$\begin{aligned}\psi_{1|2}(\tau) &= \langle 0|\hat{a}_1(\tau)|\Psi_1\rangle_2|_{t_2=0} \\ &= \frac{1}{2\pi\sqrt{R}} \int d\Omega \Phi(\Omega) e^{-i(\omega_{10}+\Omega)\tau} \\ &= \psi_0(\tau) e^{-i\omega_{10}\tau},\end{aligned}\quad (11)$$

where $\psi_0(\tau) = \frac{1}{2\pi\sqrt{R}} \int d\Omega \Phi(\Omega) e^{-i\Omega\tau}$ is the Fourier transform of the two-photon joint spectrum function. Equation (11) also clearly demonstrates that the quantum state of the heralded single photon is a coherent superposition of the frequency components in its spectrum, but not a mixed state as commonly believed. The time origin settled by the trigger photon allows for shaping the heralded single photons with an electro-optic modulator [23]. The recent experiments of single photons coherently interacting with atoms [24–26] and cavities [27, 28] have

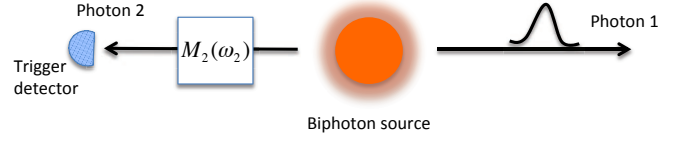


FIG. 2. (color online). Nonlocal modulation. A spectrum modulator $M_2(\omega_2)$ is placed on the path of the trigger photon 2 to nonlocally shape the heralded photon 1.

strongly evidenced that these heralded narrowband single photons indeed have coherent wave packets, but their quantum-state purities have never been formally and theoretically justified.

Following Eqs. (4) and (11), we can produce many interesting heralded single-photon states by engineering the biphoton joint spectrum and frequency entanglement. For example, from a frequency-bin entangled two-photon state $|\omega_{10} + \delta\rangle|\omega_{20}\rangle + e^{i\theta}|\omega_{10}\rangle|\omega_{20} + \delta\rangle$, one can generate a heralded single-photon two-color qubit state $|\omega_{20}\rangle + e^{i\theta}|\omega_{20} + \delta\rangle$. This will certainly find important applications in quantum information processing and quantum communication.

As compared to the heralded photons generated from an frequency-uncorrelated source, heralding single photons from the frequency-entangled (anti-correlated) biphotons has two unique features and advantages. The first is that we can reverse the relative time by switching the trigger photon. In the above discussion, we take photon 2 as the trigger photon and obtain the heralded photon 1 with a temporal waveform $\psi_{1|2}(\tau) = \psi_0(\tau)e^{-i\omega_{10}\tau}$ given in Eq. (11). Now let's switch the trigger detection to photon 1 to heralding the presence of photon 2. We obtain the temporal wave function of the heralded photon 2 as $\psi_{2|1}(\tau) = \psi_0(-\tau)e^{-i\omega_{20}\tau}$, whose amplitude envelope is the time-reversal of the heralded photon 1. This time reversal feature, resulting directly from the frequency anti-correlation, have an important application. For example, when generated from an atomic multi-level system, the biphoton correlation can exhibit exponential decay waveforms because of the nature of spontaneous emission [20]. Depending on which photon is detected as the trigger, we can generated heralded photons with an exponential growth or decay waveform that are particularly useful for loaded them into a cavity or interacting them with atoms [26, 27]. The second interesting feature is the nonlocal spectrum modulation [29–31], which allows for shaping the heralded photon by placing a spectrum modulator $M_2(\omega_2)$ on the trigger photon path, as illustrated in Fig. 2. It can be derived that, the temporal wave function of the heralded photon becomes

$$\psi_{1|2}(\tau) = \frac{1}{2\pi\sqrt{R}} \int d\Omega \Phi(\Omega) M_2(\omega_{20} - \Omega) e^{-i(\omega_{10}+\Omega)\tau}.\quad (12)$$

If the photon 1 experiences any distortion caused by

the propagation dispersion, it can also be nonlocally corrected by placing a dispersion compensation element on the path of the trigger photon 2.

In summary, we demonstrate that the detection of one photon from a frequency-entangled (frequency-anti-correlated) two-photon state, as the time origin established by the trigger photon detection has uncertainty much shorter than the two-photon coherence time, projects the remaining photon onto a pure single-photon state. The physics behind such a heralding process can be pictured as the following: the trigger detection erases the frequency information of the trigger photon, and as a result the heralded photon is in a superposition state of its frequency components. For the first time, we provide a full theoretical justification on the quantum-state purity of such heralded photons, while they had been commonly believed as mixtures of multiple frequency components. We show that the quantum-state purity depends on the response time of the trigger-photon detector. A purity of higher than 0.98 can be achieved as the trigger-detection response time is shorter than one-tenth of the biphoton coherence time (inverse of the bandwidth). The purity becomes significantly degraded as the trigger-detection response time is longer than the biphoton coherence time. We also show that such a heralded narrowband single photon has a well-defined coherent wave packet. The entanglement raised from the frequency anti-correlation allows for reversing the relative time by switching the trigger photon and shaping the heralded single photon through nonlocal spectral modulation.

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